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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

ROLF TITTMANN

SERIAL NO. 10/529,408

FILED: March 29, 2005

FOR: OPAQUE, COLOUR-PIGMENTED COATINGS

Group Art Unit: 1796

Examiner: KRIELLION A. SANDERS

Commissioner for Patents
Washington, D.C. 20231

DECLARATION UNDER 37 CFR 1.132

I, Roman Lenz, a citizen of Switzerland, residing in 4410 Liestal, Switzerland, hereby declare:

That the University of Basel, Switzerland, awarded me the degree of Doctor of Sciences (chemistry) in 1996;

That I have 10 years of experience in the field of organic dyestuffs and pigments, especially in the synthesis and screening of organic pigments;

That for 10 years I have been in charge of a lab team dealing with polycyclic pigments, especially of the DPP type, for the coloration of high molecular weight organic materials within CIBA, Basle, Switzerland;

That I am the inventor or co-inventor of a number of patents, including US Patents in fields closely related to that of the above-identified application;

That I have read carefully the instant application 10/529,408 as well as US patents 5,618,343 (Hendi and Jaffe) and 5,821,373 (Hao and Wallquist), and that I am very familiar with the subject matter thereof;

That, in the past, I worked together with Tittmann, Hendi, Jaffe, Hao and Wallquist on a number of projects and know their techniques very well;

That I was asked to compare a composition according to the instant application 10/529,408 with the closest prior art, because the inventor Rolf Tittmann as well as prior art inventors Zhimin Hao and Olof Wallquist had moved to different positions outside Pigments Research & Development;

That the experiments described in the following have been carried out by me or under my supervision and the evaluation of the test results has been done by myself.

EXPERIMENTAL

Comparative sample A was made according to example 9 of US patent 5,821,373.

Sample B according to the invention was made by mixing 3,6-bis(4-biphenyl)-2,5-dihydropyrrolo[4,3c]pyrrole-1,4-dione (C.I. Pigment Red 264) of specific surface area ~ 25 m²/g with 3-phenyl-6-(4-biphenyl)-2,5-dihydropyrrolo[4,3c]pyrrole-1,4-dione of specific surface area ~ 10 m²/g, in the same proportions as in example 9 of US patent 5,821,373, without any further treatment of the physical mixture.

Samples A and B were incorporated in usual manner into a paint system analogous to that of example 14 of US patent 5,821,373, comprising 5% of pigment, 66% of cellulose acetobutyrate solution, 24.4% of Dynapol® H-700-08, 3% of Maprenal® MF650 and 1.6% of Disperbyk® 160 (all % by weight). The final dispersion was bar-coated on black and white paper.

The CIE 1976 L*C*h color space values of the coatings were measured according to standard test procedures. The results were as follows:

Sample	L*	C*	h	L*	C*	h	ΔE^*
	over white			over black			
A (comparative)	30.4	29.2	19.4	26.1	7.5	20.2	22.3
B (instant)	37.4	47.1	23.4	34.9	37.8	23.2	10.1

According to usual best practice in the art, the L^*C^*h values were measured over white and over black and the ΔE^* value was then calculated as the color difference between the L^*C^*h values over white and those over black. Perfectly transparent samples would show values of zero for each of L^* and C^* over black, leading to extremely high ΔE^* values, while perfectly opaque samples would show the same L^*C^*h values over black and over white, leading to a ΔE^* value of zero.

The coating comprising instant sample B had a much lower ΔE^* value than the comparative sample A. This reflects the fact that instant sample B was much more opaque than comparative sample A, which fact was already visually apparent on the coatings. As compared with the coating comprising comparative sample A, the coating comprising instant sample B also has higher lightness (L^*) and much higher chroma (C^*) values, especially over black, as well as a significantly different hue (h).

DISCUSSION OF RESULTS

The results are in line with the skilled artisan's knowledge. It is well-known in the art that the presence of guest molecules in a host pigment crystal lattice leads to distortions of the crystal lattice, thus to unpredictable, different shades. US patent 5,821,373 indeed discloses such shifts in shade, which are further said not to adversely affect the good pigment properties (column 1 / lines 36-40). This implies that the pigment particles are essentially similar to those of pure Pigment Red 264. However, a higher opacity is not expected.

Moreover, the invention relies on the physical combination of two different kinds of pigment particles, each having its own selected specified surface area, to improve the hiding power of the first component. In a single-phase solid solution such as disclosed in US patent 5,821,373, however, the guest pigment is not present as sizeable particles, but as isolated molecules embedded in the crystal lattice of the host (it is explicitly disclosed that the crystal lattice is identical with that of the host). This modifies of course the light absorption characteristics, thus the color, but has little influence on the refractive index, thus reflection and scattering. The hiding power of pigments depends on all physical parameters of its crystals.

DISCUSSION OF US PATENT 5,618,343 (HENDI AND JAFFE)

US patent 5,618,343 (Hendi and Jaffe) discloses pigment compositions useful to prepare pigmented coatings that exhibit geometric metamerism or metallic flop, and comprising a small particle size pigment and a flop-enhancing agent (abstract).

However, it is important to consider the whole teachings of US patent 5,618,343, which comprise a few more essential points:

- An effect pigment is comprised as a mandatory component (claims 1 and 22).
- The small particle size pigments are transparent and they lack light scattering (column 3 / lines 65-67 and column 6 / lines 37-39). There is no intention at all to get any opacity from the color pigments. On the contrary, reflectivity relies only on the additional presence of metallic or interference pigments (examples 14, 16 and 18). An excellent distinctness of image is obtained, too.
- The small particle size pigments are treated with the flop-enhancing agents (column 1 / lines 13-19) in order to enhance the flop of coatings. In other words, flop is not created, but only the already present flop is enhanced. Flop is only visible in combination with effect pigments (column 7 / lines 21-29). In any case, the color pigment is of course required to be transparent as mentioned above.
- The flop-enhancing agents must be copper phthalocyanine, indanthrone or carbazole dioxazine compounds, which are green, blue or bluish violet. In all examples with the exception of example 5, at least partially soluble derivatives of these pigment classes are used as flop-enhancing agents. It is clear that these soluble derivatives adsorb on the surface of the small particle size, transparent pigments. In example 5, both pigments are intimately mixed then pulverised together in a preliminary step, leading to aggregation of both pigments, too.
- There is no disclosure or suggestion of any specific surface area for the flop-enhancing agent. Obviously, the flop-enhancing agent's particle size is considered either to be irrelevant^(†), or at least not different from that of the small particle size, transparent pigment.

(†) Possibly true for phthalocyanines, which do not significantly scatter independently of their particle size.

In summary, US patent 5,618,343 discloses transparent color pigment particles in combination with opaque flop agents, such as aluminum or interference mica flakes, and additionally flop enhancing agents. The transparency of the color pigment is essential. The skilled artisan knows that opaque color pigments would scatter light and obscure the effect pigments, thus reducing or even annihilating the flop, which would be in total contradiction with the goal of US patent 5,618,343.

The skilled artisan further knows, that the opacity of a pigment does not depend solely on its specific surface area. On the contrary, it depends largely not only on the particle size, but also on the particle size distribution as well as the shape and the refractive index of the particles. The latter three parameters depend themselves on the pigment's chemical identity. US patent 5,618,343 specifies a transparent pigment, on one hand including Pigment Red 264 (amongst many others), and on the other hand having a specific surface area of 40–100 m²/g (broadest range). However, this cannot be construed to disclose the instant Pigment Red 264 of specific surface area 20–50 m²/g (preferably 30–40 m²/g), which is clearly opaque in full agreement with my own experience. Pigment Red 264 does not belong to the few pigments of specific surface area ≤ 50 m²/g which are transparent, as required in US patent 5,618,343. Opaque Pigment Red 264 of specific surface area 20–50 m²/g (preferably 30–40 m²/g) is evidently neither taught nor suggested in US patent 5,618,343.

Hence, US patent 5,618,343, wherein transparent color pigments are used and the opacity of the coating results from the use of aluminum or interference mica pigments, pertains to a technology which is fundamentally different from that of the instant application 10/529,408, wherein the hiding power results from the opacity of the color pigments themselves. Instantly, a color effect appears only optionally, when the color pigments are applied in adjacent layers instead of both together in the same layer (see example 2). And even in this particular case, the color effect is due to a physical principle which is fundamentally different from that of US patent 5,618,343: perpendicular light just penetrates deeper and is reflected by both sublayers, while oblique light is reflected mainly by the top layer.

CONCLUSIONS

Above experimental comparison with the closest prior art showed that an instant physical mixture of two pigments having each a selected specific surface area is much superior in opacity, as compared with a solid solution of same chemical composition according to example 9 of US patent 5,821,373 (Hao and Wallquist).

The skilled artisan would not consider US patent 5,618,343 (Hendi and Jaffe) as an adequate starting basis for improving the opacity of a red pigmented coating. On the contrary, US patent 5,618,343 definitely requires transparent color pigments.

Moreover, even if the skilled artisan would nevertheless consider both US patents 5,821,373 and 5,618,343, he would not be able to combine their teachings to get the instant invention of Tittmann (serial number 10/529,408). For the skilled artisan, the only meaningful combination of US patents 5,618,343 and 5,821,373 would be to use a solid solution according to US patent 5,821,373 as the transparent color pigment of specific surface area of 40–100 m²/g in the flop composition of US patent 5,618,343. However, one would not deviate from the essential teaching of US patent 5,618,343, that the color pigment must be transparent, nor would one dismiss the flop producing and the flop enhancing agents. Therefore, the skilled artisan would not have obtained the instant opaque coatings.

Hence, US patents 5,618,343 (Hendi and Jaffe) and 5,821,373 (Hao and Wallquist), either alone or in combination with each other, fail to disclose or suggest the instant invention, which is therefore clearly novel and not obvious.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false

statements may jeopardize the validity of this application or any patent issuing thereon.

Signed, this 10th day of March, 2008



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Production, Properties, Applications



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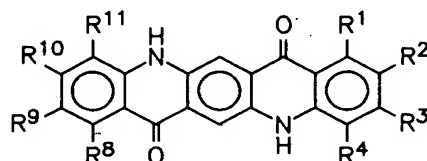
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Representative commercial quinacridone pigments are listed in Table 28. Each specimen is offered in only one crystal modification. Unsubstituted quinacridone is somewhat of an exception in that two crystal phases are commercially available.

Tab. 28: Commercially available quinacridone pigments.



C.I. Name	C.I. Constitution No.	R ²	R ³	R ⁴	R ⁹	R ¹⁰	R ¹¹	Range of shades	Total number of known modifications
Quinacridone pigments									
P.V.19	73900	H	H	H	H	H	H	red-violet to bluish red	5
P.R.122	73915	CH ₃	H	H	CH ₃	H	H	bluish red (magenta)	4
P.R.192	-	-	-	-	-	-	-	bluish red	1
P.R.202	73907	Cl	H	H	Cl	H	H	bluish red to violet	3
P.R.207	73906	H	H	Cl	H	H	Cl	yellowish red	1 (4,11-dichloro:3)
P.R.209	73905	H	Cl	H	H	Cl	H	red (mixed with 1,8- and 1,10-dichloroquinacridone)	1
Quinacridone quinone pigments (Quinacridone quinone, C.I. 73920)									
P.R.206	-	H	H	H	H	H	H	maroon (golden yellow)	1
-	-	H	H	Cl	H	H	C	scarlet	1 (4,11-dichloro:3)
P.O.48	-	unsubstituted quinacridone + quinacridone quinone						maroon (golden yellow)	1
P.O.49	-	unsubstituted quinacridone + quinacridone quinone						maroon (golden yellow)	1
P.V.42	-							maroon	1

Individual Pigments

Pigment Violet 19, β -Modification

The β -modification affords a reddish violet shade. These pigments are frequently used in combination with inorganic pigments, especially with iron oxide, or to a decreasing extent with Molybdate Red pigments, to color industrial paints. Such pigment combinations provide comparatively dull shades of red to bordeaux. The excellent hiding power of the resulting systems is attributed to the content of highly

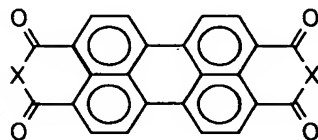
areas as well as high hiding power. Types with fine particle sizes are used especially in metallic and transparent paints, while opaque types provide full shades, frequently in combination with inorganic or other organic pigments. Some types are particularly suited to use in plastics and in spin dyeing products, in which they demonstrate excellent heat stability. However, perylene pigments are rarely used to color HALS (Hindered Amine Light Stabilizer) stabilized polyolefins, i.e., polyolefins that are stabilized by steric amines. At medium to high pigment concentrations, these stabilizers may be inactivated or even destroyed by exposure to light, rapidly converting the polyolefin system into a brittle material.

3.4.1.5 Commercially Available Perylene Pigments

General

Representative commercial perylene pigments are listed in Table 29:

Tab. 29: Commercially available perylene pigments.



C.I. Name	C.I. Constitution No.	X	Shade
P.R.123	71145		scarlet to red
P.R.149	71137		red
P.R.178	71155		red [5]
P.R.179	71130		red to maroon
P.R.190	71140		bluish red
P.R.224	71127		bluish red
P.V.29	71129		red to bordeaux
P.Bl.31	71132		black
P.Bl.32	71133		black